

**MOBILE TERAWATT FEMTOSECOND LASER SYSTEM (MTFLS) FOR LONG  
RANGE SPECTRAL SENSING AND IDENTIFICATION OF BIOAEROSOLS AND  
CHEMICAL AGENTS IN THE ATMOSPHERE**

5 This patent application claims the benefit of U.S. Patent Application Serial No. 60/458,757, filed March 28, 2003 which is hereby fully incorporated by reference.

**FIELD OF THE INVENTION**

10 The present invention is related to detection and identification of airborne biological and chemical threats in real time at distances from a few meters to several kilometers. Advanced compact femtosecond terawatt laser technology is combined with state-of-the-art spectroscopic and computational methods. These methods are implemented in a unique mobile standoff detection system.

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**BACKGROUND OF THE INVENTION**

US Pat. No. 5,175,664 to Diels et al. describes a method and arrangement for discharge of lightning using ultra-short laser pulses. This proposed method enables discharges of electricity transmitted via conductive ionized channels produced by one or more first laser pulses of wavelengths essentially within the ultraviolet (UV) range. The preferred wavelength of operation is about 248 nm and the pulse duration of the laser is of the order of 100 fs. In accordance with this invention, lightning is triggered by creating an ionized channel by one or more femtosecond UV pulses and simultaneously sending one or more laser pulses of longer wavelength and duration through the same path. In this way the conductivity of the laser induced channel can be maintained long enough for discharges and lightning to occur. However, this patent does not address applications associated with atmospheric spectral sensing of atmospheric gases, pollutants and biological agents using femtosecond terawatt laser generated filaments as light sources.

30 One other related reference is U.S Pat. No. 5,726,855 to Mourou et al., entitled "Apparatus and Method for Enabling the Creation of Multiple Extended Conduction Paths in the Atmosphere" which claims an apparatus and method for the creation of multiple extended conduction paths in the atmosphere using high peak-power ultra-short laser pulses. Furthermore, the same group

published addressing long range self-channeling of intense femtosecond pulses in air in 1994 by Mourou and co-workers. The focus of their patent is centered on an apparatus for controlling the discharge of lightning strikes and grounding means using a grounding tower. Also no claims are made regarding spectroscopic optical sensing and identification of bio-agents using multiple extended conduction paths in the atmosphere.

It will therefore be appreciated that there remains a need for an improved system and technique for determining whether certain molecules, such as bioaerosols and chemical agents, may be found within a sample.

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### SUMMARY OF THE INVENTION

Early warning of biological attack and plume prediction for biological aerosols require the capability to detect bio-agents over large distances. Thus, the present invention relates to a complex bio-agent sensing system based on the principle of using femtosecond terawatt lasers, preferentially titanium sapphire laser systems in conjunction with chirp-pulse amplification and compression. The propagation of such ultra-short laser pulses gives rise to strongly non-linear optical processes in the atmosphere, leading to filamentation of the laser pulse at a specific distance. Conducting plasma channel produce a white-light supercontinuum spanning the ultraviolet (UV), visible (VIS) and infrared (IR). Wavelength region. This novel atmospheric lamp can be used for spectroscopic analysis of bio-agents by means of a Lidar (Light Detection and Ranging) system. The supercontinuum can be generated directly in a particle cloud and hence is suitable for multi-spectral long-range detection and identification of bio-aerosols as well as chemical pollutants and radioactive isotopes. According to the invention, compact femtosecond terawatt laser technology is applied in combination of state-of-the-art spectroscopic and computational methods for spectral sensing of bio-aerosols with molecular specificity.

The current invention permits real time detection, discrimination, and identification of the full spectrum of threats including; toxins, spores, bacteria and virus. The sensing range spans up to 10,000 meters and can be applied to airborne versions together with stand alone trigger devices at strategically vital locations on sea, air or land.

This invention presents a promising new trigger sensor and a standoff detection system that can identify airborne biological, chemical, and nuclear agents within a few seconds at distances from a few meters to several kilometers. The biological agent-sensing device is based on the principle of remote differential time resolved monitoring of the atmosphere using 5 ultrashort terawatt laser pulses. According to this invention, concept of advanced compact femtosecond Terawatt (fs-TW) laser technology is combined with state-of-the-art spectroscopic and computational methods. The stand off sensing technology permits real time detection, discrimination, and identification of the full spectrum of threats including; toxins, spores, bacteria and virus. The sensing range spans up to 10,000 meters and can be applied to 10 airborne versions together with stand alone devices at strategically vital locations on sea, air or land. This system would reduce the probability of false alarm and time for detection by more than one order of magnitude. The minimum anticipated range would be of the order of 5-10 spores per liter of air.

15 In one embodiment, the invention is provided in a system for determining the constituents of a sample, the system comprising, a femtosecond terawatt laser radiation source configured to emit laser radiation through a sample, an optical unit configured to receive light backscattered from the sample, and a detection and analysis unit coupled to the optical unit for analyzing a spectral signature of the sample. The system may also include an optical fiber cable coupling the optical unit to 20 the detection and analysis unit.

In some variations, the detection and analysis unit comprises an integrated diagnostic unit having one or more infrared and UV/VIS spectrometers with gated detection capability, two photomultipliers attached to an air transient digitizer, and a data acquisition control unit. In addition, or 25 in the alternative, the detection and analysis unit may further comprise a real-time computing system for identification and discrimination of at least one of the group comprising aerosols, airborne bacteria, viruses, toxins, dust particles, pollen, water droplets, gaseous agents, and pollutants.

30 The femtosecond terawatt laser radiation source may be amplified by a variety of techniques such as chirped pulse amplification. It may also be, in some examples, a laser such as a Ti:Sapphire laser that is configured to emit energy of approximately 300 mJ per pulse. The femtosecond

terawatt laser radiation source may have a pulse power of about approximately 3 and 4 TW with a pulse duration approximately of the order of 80 to 100 fs and a repetition rate of approximately 10 Hz. It may emit light within a spectral range approximately centered at 800 nm or 267 nm with a spectral width of approximately 20nm. In addition, the femtosecond laser radiation source may  
5 emits laser pulses at a center wavelength of approximately 800nm and spectral width of 20 nm to create plasma filaments as well as at a wavelength of approximately 267nm.

The detection and analysis unit may configured to detect airborne biological, chemical agents and water droplets by at least one technique chosen from the group comprising: differential  
10 absorption, Raman Raleigh and Mie scattering, fluorescence, floorescence LIDAR measurements, ground-based LIDAR measurements, air-based LIDAR measurements, and Raman LIDAR measurements. The detection and analysis unit may also be configured to provided 3D maps or other representations of detected molecules to further assist with their identification.

15 In another variation, the invention is embodied in a method for determining the constituents within a sample, the method comprising the steps of providing a femtosecond terawatt laser radiation source configured to emit laser radiation through a sample, capturing light backscattered from the sample, and analyzing a spectral signature of the sample to determine its constituents.

20 The analyzing step may determine whether the constituents include least one of the group comprising: aerosols, airborne bacteria, viruses, toxins, dust particles, pollen, water droplets, gaseous agents, and pollutants.

25 The method may also include the step of amplifying the femtosecond terawatt laser radiation source using chirped pulse amplification.

In some variations, the femtosecond terawatt laser radiation source is a laser such as a Ti:Sapphire laser configured to emit energy of approximately 300 mJ per pulse. The method may also include the step of pulsing the femtosecond terawatt laser radiation source at a power of about  
30 approximately 3 and 4 TW with a pulse duration approximately of the order of 80 to 100 fs and a repetition rate of approximately 10 Hz. The femtosecond terawatt laser radiation source may emit

light within a spectral range approximately centered at 800 nm or 267 nm with a spectral width of approximately 20nm.

5        Optionally, the femtosecond laser radiation source may emit laser pulses at a center wavelength of approximately 800nm and spectral width of 20 nm to create plasma filaments as well as at a wavelength of approximately 267nm.

10      Depending on the desired implementation, the analyzing step may use at least one technique chosen from the group comprising: differential absorption, Raman Raleigh and Mie scattering, fluorescence, flourescence LIDAR measurements, ground-based LIDAR measurements, air-based LIDAR measurements, and Raman LIDAR measurements for determining which constituents are present within a sample.

15      The method may also include the step of generating a 3D map or image of the detected constituents. The constituents may be determined, in part, by the step of comparing at least one of detected vibrational bands, detected Raman spectra, and fluorescence spectra, with previously measured spectral data to identify the constituents within the sample.

20      A mobile femtosecond terawatt LIDAR system and/or trigger sensor for long-range analysis with high temporal and spatial resolution and spectroscopic identification of airborne biological and chemical agents is also provided. Such a system includes a femtosecond terawatt laser radiation source based on the principle of chirped pulse amplification (CPA), preferably a Ti:Sapphire laser system having an energy of approximately 300 mJ per pulse resulting in a pulse power of about 3 and 4 TW with a pulse duration of the order of 80 to 100 fs and a repetition rate of 10 Hz. The 25     laser source emits light in the spectral range of approximately 800 nm with a spectral width of approximately 20 nm or laser pulses with a wavelength of about 267 nm corresponding to third harmonic generation (SHG). The system also includes an optical system coupled to the laser radiation source including sending and receiving telescopes, an integrated diagnostic system embracing infrared and UV/VIS spectrometers with gated detection capability, two photomultipliers attached to air transient digitizer and data acquisition control system, and a real-time computing system (using techniques such as neural networks, fuzzy logic and other advanced 30     computational techniques) for identification and discrimination of at least one of the group

comprising: aerosols, airborne bacteria, viruses, toxins, dust particles, pollen, water droplets, gaseous agents, and pollutants.

The system may provide for remote monitoring of the atmosphere using ultra-short terawatt laser pulses giving rise to strongly nonlinear optical processes in the air thus generating plasma channel type of filaments encompassing: NIR femtosecond laser pulses at a center wavelength of approximately 800nm and spectral width of 20 nm creating plasma filaments caused by self-channeling/guiding effects and producing a supercontinuum atmospheric light source ranging for optical sensing applications from the UV to the MIR and UV femtosecond laser pulses operating at a wavelength of approximately 267nm corresponding to the SHG creating no new wavelengths but propagating with much lower losses up to several km through the plasma channel when compared to the NIR laser pulses.

Femtosecond laser pulses centered at 800nm may be used to produce a broadband supercontinuum light source in the atmosphere for sensing of airborne biological, chemical agents and water droplets by means of differential absorption and/or fluorescence LIDAR measurements. In addition or in the alternative, femtosecond laser pulses centered at 267nm are used for sensing of airborne biological, chemical agents and water droplets by means of fluorescence and/or Raman LIDAR measurements. Femtosecond laser pulses centered at 800nm and 267nm may be used for sensing of airborne biological, chemical agents and water droplets by means of ground based and air based LIDAR measurements. Femtosecond laser pulses centered at 800nm and 267nm may be used for sensing of airborne biological, chemical agents and water droplets by means of ground based and air based LIDAR measurements.

The system may also compare the most significant vibrational bands (fingerprint regions) of these bio-aerosols, chemical agents and water droplets with existing laboratory spectral data to rapidly detect and identify such agents for multi-spectral long-range atmospheric agent detection of clouds and plumes. In other variations, the system compares the most significant Raman and fluorescence spectra (fingerprint regions) of bio-aerosols, chemical agents and water droplets with existing laboratory spectral data to rapidly detect and identify such agents for multi-spectral long-range atmospheric agent detection of clouds and plumes.

One aspect of the invention is that it provides high range resolution over long distances to record the signal of the backscattered light due to Raleigh and Mie scattering from gaseous molecules, bio-aerosols and water droplets in the clouds and plumes. It combines the advantages of the remote sensing techniques and their broadband spectral resolution, with 3D mapping capability.

5 This permits the simultaneous measurement of several bio-aerosol compounds, even with overlapping spectral signatures.

Furthermore, the invention provides sophisticated differential absorption, fluorescence, Raman Raleigh and Mie scattering, multi-channel, multi-wavelength, multi-spectral LIDAR system for

10 ground based and air based monitoring of bio-aerosols and chemical agents in real time with high accuracy and reduced false alarms for long range detection from about a few meters to 10 km.

The invention may also be embodied in a complex trigger sensor/standoff system with substantially improved performance requirements for probability of detection, probability of false alarm, time for detection and threat level when compared to existing technologies. The invention

15 may be used in a variety of settings including water treatment plants, environmentally sensitive areas e.g. subways, airports, government buildings, military facilities, industrial complexes and cities as mobile trigger points, area sensors and standoff systems.

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#### BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a diagrammatic view of the femtosecond LIDAR system for long-range detection of bio-aerosols;

25 FIG. 2 is a schematic view of different spectroscopic excitation and de-excitation channels in the infrared and ultraviolet including scattering processes;

FIG. 3 is a bacillus subtilis extinction aerosol spectrum in the MIR and far IR region;

FIG. 4 is a bacillus subtilis absorption aerosol spectrum in the MIR region;

FIG. 5 is an example of a mobile ground based standoff LIDAR system useful for understanding and implementing the invention;

30 FIG. 6 is an example of a mobile air-based LIDAR system useful for understanding and implementing the invention;

FIG. 7 is a schematic diagram illustrating a system embodiment of the invention; and

FIG. 8 is a process flow diagram illustrating a method embodiment of the invention.

#### DETAILED DESCRIPTION OF THE INVENTION:

5 According to this invention, an fs-TW laser system is comprised of a femtosecond laser (10), pulse stretcher (11), pulse amplifier (12) and chirp generator (13). The final pulse duration is approximately 80 to 100 fs and the energy is of the order 300 mJ per pulse resulting in a pulse power between approximately 3 and 3.5 TW. The pulse shape is analyzed by a pulse diagnostic system (14) and then directed to an off-axis telescope (15). The slightly focused laser beam (16)  
10 then creates a channel (filament) in the atmosphere (17) and the emitted light from the filament interacts with the bio-aerosol cloud (18). The backscattered light (19) is collected by means of a receiving telescope (20) and then analyzed by different types of spectrometers (21 and 22) and photo-multipliers (23 and 24), where the IR spectrometer (21) measures the differential absorption in the bio-cloud and the VIS/UV spectrometer (22), the absorption and/or fluorescence  
15 of the cloud. As can be seen in Fig.1, the photo-multiplier signals (23 24) are fed to transient digitizer (25). Finally the data acquisition and control system (26) receives signals from the different detection devices.

20 The diagrammatic view (100) of Fig.1 represents a standoff laser system with a complex integrated detection setup that can be used as a biological chemical and environmental agent monitoring arrangement. This system is based on the principle of remote sensing and monitoring of the atmosphere using ultra short terawatt laser pulses. The propagation of these laser pulses gives rise to strong nonlinear optical processes in air maybe producing filamentation type of conducting plasma channels where in-turn a white-light supercontinuum (16) is created. This  
25 supercontinuum can be located directly in a bio-pathogen cloud (24), and its wavelength extends from the ultraviolet (UV) to the infrared (IR).

30 The present invention relates to currently available fs-TW laser systems based on the chirped pulse amplification (CPA) technique using Titanium: Sapphire as the active laser crystal. These lasers provide a broadband spectral distribution of light centered at a wavelength of about 800 nm.

The aim of the invention is detecting and specifying bio-aerosols using different approaches such as: (a) detection through differential absorption and UV fluorescence and (b) detection through modification of the optical characteristics of the plasma channels generated in air at different locations. In either case, light emitted in the near-backward region from an aerosol source is spectroscopically and temporally analyzed in the vicinity of the laser beam propagation. The receiver systems are capable of yielding spectra from the UV through the VIS into the MIR region (from about 270 to 5500 nm). Field test experiments will be used for different types of aerosols including water droplets, inorganic atmospheric aerosol (ammonium sulphate), common non-biological organic atmospheric aerosol (i.e., organic carbon), common biological atmospheric aerosol (e.g., pollen), airborne bacteria, viruses, toxins, dust particles, pollen, water droplets, diesel dust, gaseous agents and other biological aerosol serving as surrogate.

An object of this invention is complex plume prediction schemes and early warning systems for biological attacks. As shown in schematic (200) of Fig. 2, an fs-TW laser (27) creates a two-colored filament (28) at a specific location in the atmosphere with the wavelength of approximately 800nm, where the laser pulses interact with specific aerosols at different distances (29). As indicated in Fig. 2 the third harmonic generation (THG) component of the laser propagating inside the filament channel (28) can also be used as a new diagnostic technique. Different strategies are used to understand and interpret complex threats/attacks, namely due to simultaneously analysis of infrared differential absorption channels (30) and UV and visible channel fluorescence (31) produced by the long distance UV filaments created by 800nm laser pulses. In addition, light scattering channels including Rayleigh, Mie and Raman scattering (32) will be observed to shed more light on the composition of different atmospheric constituents and background radiation.

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The invention may also be used to study and identify micro-droplets, which represent a major part of atmospheric aerosols. They are attractive systems for the study of several nonlinear optical effects using fs-TW laser devices. They may act as lenses focusing the incident radiation onto some small regions inside the droplets and they also induce morphology dependent resonances, which can further enhance the laser intensity in the droplet. Hence, at the areas of high laser intensity the efficiency for nonlinear optical processes is strongly enhanced. The scattered wave

and the internal intensity distribution depend on the refractive index of the droplet medium and on the size parameter, which is the ratio of the droplet circumference to the wavelength of the incident light. When studying the interaction of femtosecond laser pulses with these micro-droplets, the large spectral bandwidth of the ultra short pulses has to be taken into account.

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Another object of the present invention is to utilize vibrational spectroscopy of bio-agents. This approach includes extinction and scattering measurements to characterize the spectral fingerprints and change of conformation of the biological molecular systems. Therefore, the aim of this invention also is to compare the most significant vibrational bands (fingerprint regions) of such aerosols with existing laboratory spectral data to rapidly detect and identify such bio-agents. As described in **Fig 3**, a characteristic IR spectrum (300) of aerosolized *Bacillus subtilis* var. *niger* (BG) spores in the 2.3-to 12- $\mu\text{m}$  wavelength regions is shown. The bio-agent spectrum is composed of an absorption part superimposed on a Mie scattering background (33). The absorption spectrum exhibits specific spectral features at approximately 2.9-3.6 $\mu\text{m}$  (34). The spectral peak at about 3.1 $\mu\text{m}$  can be partially identified as the Amide A band. Moreover, the spectral features located in the 5.5 to 6.6 $\mu\text{m}$  region are associated with the Amide I and Amide II bands (35). Additional important fingerprint regions occur in the range 6.6 - 8 $\mu\text{m}$  (36). Moreover spectral features between 8 – 11  $\mu\text{m}$  can be associated with Amide III band, phosphate groups and peptide backbone structures (37). In order to extract the most important spectral features (peak positions, widths, band structures, intensity ratios etc.), the noise associated with the spectral data has to be removed. Then an appropriate non-linear function due to Mie scattering will be used to fit the residual background following a peak fitting analysis. A typical resulting absorption spectrum (400) is displayed in **Fig. 4** between 3 - 4 $\mu\text{m}$ . Two different cases are considered here. In the first case, two substantially different atmospheric transmission channels (38 and 39) with similar absorption cross sections are examined. One can see here that there is a valley at the channel with lower atmospheric transmission (38) and a peak is achieved at the channel with higher atmospheric transmission (39). In the second case, constant atmospheric transmissions are considered although the absorption cross sections differ substantially. For example, channel (40) has a higher absorption cross section than channel (41) with a lower absorption cross-section. These patterns comprise the vibrational characters of the constituents such as DNA/RNA, proteins and cell wall components. Owing to the multitude of cellular components, broad and superimposed spectral bands are observed in the MIR range.

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Another example (500) useful for understanding and implementing the invention is shown in **Fig. 5**. A schematic view of a ground based differential Mobile Multiple Wavelength Trigger Sensor system and/or Teramobile Lidar system is exhibited housed in a single mobile container (42). The setup consists of a tabletop terawatt femtosecond laser (43) and detection system with high spatial, spectral, and time resolution. The system is comprised of sending and receiving telescopes (44) with the vertical beam direction (45) and a horizontal beam direction (46). The mobile container also has additional monitoring telescopes for trigger sensors (47). The reflected and scattered light is fed to the different UV, VIS, NIR and MIR spectrometers and associated detection systems (48). For the amplification the system also houses control boards (49) and power supplies (50). For the data interpretational and computational purposes the system has an advanced data acquisition control system with several attached parallel computers (51) for real time data reduction, modeling and discrimination. The temperature conditions are controlled inside the mobile container with the help of air-cooling system (52). This system is driven with a power generator (53), which is placed separately outside the mobile container to minimize the evasive effect from vibrations.

In **Fig. 6** an up looking airborne differential absorption femtosecond terawatt laser system (600) is shown. Such an airborne LIDAR system would be capable of a broad range of high priority measurements for use in an aircraft. This system could also be operated looking downwards from the aircraft compared with the system looking upwards from the ground. This improvement makes the LIDAR system an attractive airborne tool for both daytime and nighttime conditions. In the schematic diagram, a femtosecond terawatt laser (54) with adaptive optical elements (55) direct the laser beam towards a bio-aerosol cloud (56) and backscattered light (57) is collected by a telescope mirror (58) and directed via an optical fiber cable (59) to time resolved spectrometer setup (60). The airplane (61) contains all the necessary power supplies, electronics, data acquisition control systems and parallel computers for real time modeling and discrimination.

The main advantage of this ultrafast ground based and air –based Lidar technique over other remote sensing methods (such as differential optical absorption spectroscopy, Fourier transform infrared spectroscopy and satellite based spectroscopy) is the high resolution over long distances, which is achieved by the use of short-pulse lasers and fast detection systems to record the signal of the backscattered light arising from gaseous molecules and aerosols femtosecond white-light

Lidar combines the advantages of those remote sensing techniques and their broadband spectral resolution, with 3D mapping capability. This permits the simultaneous measurement of several bio-aerosol compounds, even with overlapping spectral signatures. Moreover, since the whole spectrum can be simultaneously acquired, the laser shot-to-shot fluctuations do yield much less  
5 systematic errors as commonly in the case for traditional Differential Absorption Lidar (DIAL) systems.

Such nonlinear effects induced by ultra short, high-power laser pulses in aerosols rely either on the micro cavity behavior of spherical micro-droplets, providing strong feedback for stimulated  
10 processes, or on the internal focusing of the incident light, providing high intensity hot spots where the efficiency for nonlinear optical processes is strongly enhanced.

High-intensity laser pulses can cause water droplets to emit white light. This technique can potentially be used to analyze the composition of clouds and shed more light on how clouds may  
15 contain bio-aerosols. This approach would also provide more information on cloud-aerosol interactions. The possibility of identifying the fingerprints of chemical components inside an individual water droplet may open up new ways to diagnose aerosol clouds and more localized sources.

20 The present invention also relates to pattern recognition procedures of the data consisting of four steps: i) cluster the feature vectors from a population of entities into classes via the robust fuzzy clustering algorithm, ii) compute a prototype for each class, iii) compute an inverse covariance matrix for each prototype and center a Gaussian fuzzy set membership function on it to construct a fuzzy classifier and place it on-line to receive incoming feature vectors. The 1-sigma (standard deviation) regions under each Gaussian are ellipsoidal in N dimensions (for N features), so a class  
25 may contain one or more ellipsoidal groups.

An alternative method related to this invention is to use the clustered signatures to train the powerful new radial basis functional link net, which learns faster and more efficiently than other  
30 types of neural networks (NN's). While there are NN's that learn, or self-organize themselves,

they are relatively slow and do not learn robustly, so clustering should be done first and then supervised training should be done for the best results.

5       **FIG. 7** illustrates a schematic diagram of a system embodiment (700) of the current invention for determining the constituents of a sample (720). The system includes a femtosecond terawatt laser radiation source (710) configured to emit laser radiation through the sample (720), an optical unit (730) configured to receive light backscattered from the sample, and a detection and analysis unit (740) coupled to the optical unit for analyzing a spectral signature of the sample. This system may be used in connection with any of the above techniques and sub-systems to further enhance  
10      the detection and analysis of molecules or constituents of interest within the sample (720).

Similarly, **FIG. 8** illustrates a process flow diagram of a method embodiment (800) of the current invention useful for determining the constituents of a sample. At step (810), a femtosecond terawatt laser radiation source configured to emit laser radiation through sample is provided.  
15      Next, backscattered light from the sample is captured at step (820). The method ends, at step (830), with the analysis of the spectral signature of the sample as captured to determine its contents. As can be appreciated, the method may be implemented with any one of the above laser radiation systems as well as detection and analysis sub-systems.

20      The drawings and foregoing description are not intended to represent the only form of the invention in regard to the details of the construction and manner of operation. In fact, it will be evident to one skilled in the art that modifications and variations may be necessary without departing from the spirit and scope of the invention. Although specific terms have been employed, they are intended in generic and descriptive sense only and not for the purpose of limitation.